Two mechanisms of pseudogap formation in Bi-2201: Evidence from the c-axis magnetoresistance

A. N. Lavrov, Yoichi Ando and S. Ono

Central Research Institute of Electric Power Industry, 2-11-1 Iwato-kita, Komae, Tokyo 201-8511, Japan

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Abstract. – Measurements of the c-axis resistivity and magnetoresistance have been used to investigate the pseudogap (PG) behavior in $Bi_{2+z}Sr_{2-x-z}La_xCuO_y$ (Bi-2201) crystals at various hole densities. While the PG opening temperature T^* increases with decreasing hole doping, the magnetic-field sensitivity of the PG is found to have a very different trend: it appears at *lower* temperatures in more underdoped samples and vanishes in non-superconducting samples. These data suggest that besides the field-insensitive pseudogap emerging at T^*, a distinct one is formed above T_c as a precursor to superconductivity.

Introduction. – In high-T_c cuprates, the electronic density of states (DOS) near the Fermi energy has been demonstrated to decrease gradually with decreasing temperature, resulting in the pseudogap (PG) formation [1–4]. This PG, which has been found to progressively "destroy" the Fermi surface (FS) [4,5], is on one hand a challenge to the conventional view of the FS itself, while on the other hand it allows one to reconcile the small number of carriers that participate in the charge transport in underdoped cuprates with the large FS observed by photoemission [1,5].

Although the existence of the PG has been documented by many experiments [1], its nature and, particularly, its relation to the superconductivity (SC) remain far from being clear. For example, according to photoemission and surface-tunneling studies [2–4], the PG evolves smoothly into the SC gap below T_c, which implies a precursor-pairing origin of the PG; in contrast, recent observations of a distinct SC gap that coexists with the PG below T_c and tends to close at T_c [6,7] suggest that the PG might have nothing to do with superconductivity [8,9]. Apparently, a crucial test for the origin of the PG would be its sensitivity to the magnetic field; however, studies of the magnetic-field dependence of the PG have reported surprisingly controversial results [10–14].

While the electronic DOS in cuprates has mostly been studied by photoemission [3–5] or surface-tunneling [2,15] spectroscopies, the extremely anisotropic nature of the Bi-based cuprates, where the crystal structure itself forms a stack of tunnel junctions, offers a possibility of *intrinsic* tunneling spectroscopy [6,7,16]. In fact, it has been shown that the c-axis transport in the Bi-based cuprates is governed by the tunneling between CuO_2 planes and, accordingly,

the c-axis voltage-current characteristics directly reflect the DOS structure of the CuO_2 planes [6,7,16]. It was reported that the c-axis differential resistivity ρ_c^d at high bias shows virtually no temperature dependence, indicating that the tunneling mechanism as well as the DOS away from the Fermi level are temperature independent [6]. However, the low-bias ρ_c^d reveals a pronounced upturn upon decreasing temperature below T^*, demonstrating the opening of the PG near the Fermi energy [6,7,16]. This tunneling behavior of ρ_c^d is in sharp contrast to the metallic in-plane resistivity that is governed by scattering and is believed to decrease with the PG opening [17].

Since the conventional c-axis resistivity ρ_c corresponds to ρ_c^d at zero bias, ρ_c of the Bi-based cuprates measured above T_c reflects the DOS at the Fermi energy [6,7]. This means that ρ_c provides a convenient probe to trace the behavior of the PG, whatever its nature is. Indeed, the characteristic temperature T^* below which $\rho_c(T)$ shows a "semiconducting" upturn coincides well with the pseudogap temperatures determined by other methods [15]. Correspondingly, the c-axis magnetoresistance (MR), $\Delta\rho_c/\rho_c$, is expected to bear information on the magnetic-field dependence of the PG, and the negative c-axis MR [14,18] has been proposed to reflect a partial recovery of the DOS with magnetic field. So far, however, most of the MR studies were done on cuprates at nearly optimum doping, where T^* closely approached T_c, and thus it was difficult to distinguish whether the observed effect was related to the SC fluctuations [18,19] or to the normal-state PG [14]. Therefore, it is desirable to extend the MR study to the heavily-underdoped region where T^* and T_c are far apart.

In this Letter, we report the study of ρ_c and the c-axis MR in $Bi_2Sr_{2-x}La_xCuO_y$ (BSLCO) and $Bi_{2+z}Sr_{2-z}CuO_y$ (BSCO) single crystals for a wide doping range, from superconducting compositions with T_c of 30 K to heavily-underdoped non-superconducting ones. The evolution of $\rho_c(T)$ indicates that T^* increases with decreasing hole doping without being interrupted by the disappearance of SC. In heavily-underdoped samples, the MR appears to be extremely small in a wide temperature range below T^*, suggesting a magnetic-field-insensitive nature of the normal-state PG. However, it is found at the same time that a noticeable negative MR, indicating a recovery of the pseudogapped DOS, starts to show up upon approaching T_c. We discuss that the appearance of the negative MR well below T^* is likely to be associated with a secondary pseudogap, which can be suppressed with magnetic fields. The data therefore point to a possibility of two distinct mechanisms for the PG in cuprates: The first one is insensitive to the magnetic-field, gains strength with decreasing hole doping, and ultimately causes a transition into an insulating state; the second one is sensitive to the magnetic field and is clearly related to the superconductivity.

Experimental methods. – Single crystals with nominal compositions of $Bi_2Sr_{2-x}La_xCuO_y$ ($x = 0.2 - 1.0$) and $Bi_{2.2}Sr_{1.8}CuO_y$ are grown by the floating-zone method [20]. Substituting Sr with La reduces the hole density in $Bi_2Sr_{2-x}La_xCuO_y$; optimum doping corresponds to $x \approx 0.45$ ($T_c \approx 30$ K) and the system becomes non-SC for $x > 0.9$ (heavily underdoped). The $Bi_{2.2}Sr_{1.8}CuO_y$ crystal is nearly optimally doped (its hole density can be estimated [21] to be $\sim$17% per Cu), and yet its T_c (onset at ~ 3 K) is much smaller than in slightly underdoped BSLCO with $x = 0.5$ ($T_c = 29.7$ K; $\Delta T_c \approx 0.35$ K), whose hole density is $\sim$15% per Cu [21].

The ρ_c measurements are done on samples with typical sizes of $1 \times 1 \times 0.1\,\mathrm{mm}^3$ using the ac four-probe technique. To provide a homogeneous current flow along the c-axis, two current contacts are painted to almost completely cover the opposing ab-faces of the crystal; two voltage contacts are placed in the small windows reserved in the center of the current contacts [22]. The MR measurements are carried out by sweeping the magnetic field to ± 14 T at fixed temperatures stabilized by a capacitance sensor with an accuracy of ~ 1 mK; this method allows reliable measurements of $\Delta\rho_c/\rho_c$ as small as 10^{-5} at 10 T.

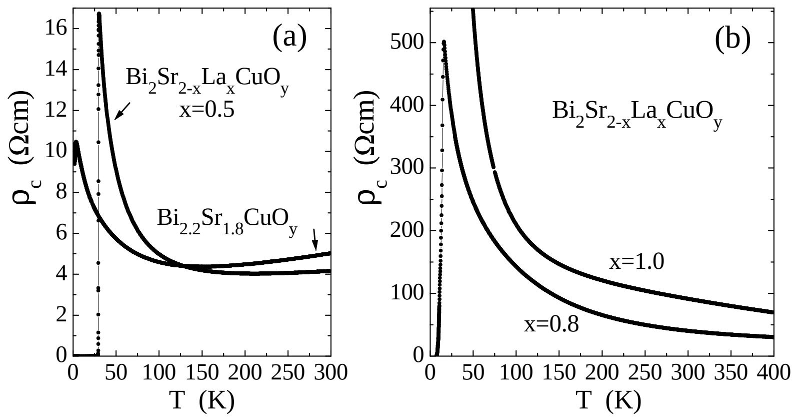

Fig. 1 – The c-axis resistivity of nearly optimally doped (a) and underdoped (b) BSLCO and BSCO single crystals. The data show the "semiconducting" low-temperature upturn, which marks the pseudogap opening.

Results and discussion. – All the Bi-2201 samples demonstrate a steep increase in $\rho_c(T)$ at low temperatures (fig. 1), which has been shown to originate from the PG formation and corresponding decrease in the DOS at the Fermi energy [6, 7, 15, 16]. One can infer that the onset of the ρ_c upturn shifts to higher temperatures with decreasing hole doping, as has been reported for other cuprates [1, 3, 8, 15], and that the evolution of the $\rho_c(T)$ behavior is apparently uninterrupted by the disappearance of SC [fig. 1(b)].

As is mentioned in the introduction, whether the PG is affected by the magnetic field or not depends crucially on its origin, and the MR data are useful in clarifying this point. For all superconducting samples we have observed that an application of the magnetic field causes a considerable decrease in the normal-state ρ_c in a rather wide temperature range above T_c;

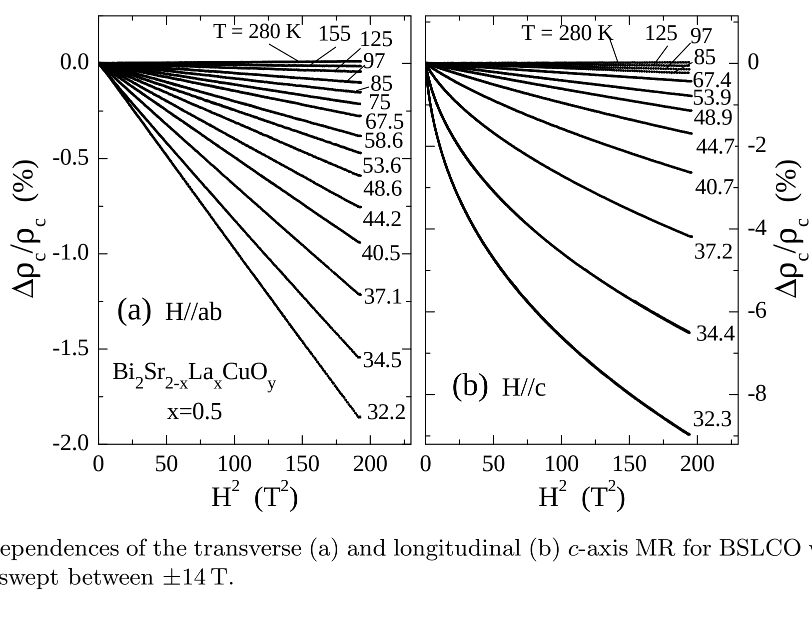

Fig. 2 – H dependences of the transverse (a) and longitudinal (b) c-axis MR for BSLCO with $x = 0.5$. The field is swept between ± 14 T.

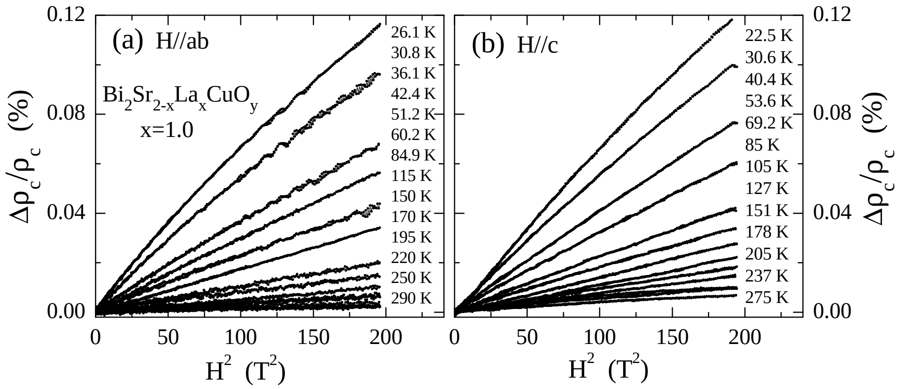

Fig. 3 – H dependences of the transverse (a) and longitudinal (b) c-axis MR for BSLCO with $x = 1.0$.

as an example, precise MR data for $x = 0.5$ are shown in fig. 2. Note that $\Delta\rho_c/\rho_c$ is strictly proportional to H^2 at all temperatures down to T_c when H is parallel to ab [fig. 2(a)], while the proportionality to H^2 is observed only at high enough temperatures when H is parallel to c [fig. 2(b)]. In the non-SC ($x = 1.0$) sample, the MR turns out to be much weaker and positive at all temperatures (fig. 3), which gives evidence for an intimate connection between the negative c-axis MR and superconductivity.

The temperature dependences of the MR for underdoped and optimally-doped crystals are depicted in fig. 4 (all the plotted data are from the region where deviation from the $\Delta\rho_c/\rho_c \propto H^2$ behavior is insignificant). It is notable that a rapid growth of the negative MR takes place with approaching T_c that decreases with decreasing hole density, rather than correlates with the upturn in $\rho_c(T)$ which shifts to higher temperatures. Intriguingly, the system seems to "know" whether it will eventually become superconducting already at $\sim 150\,\mathrm{K}$, where the

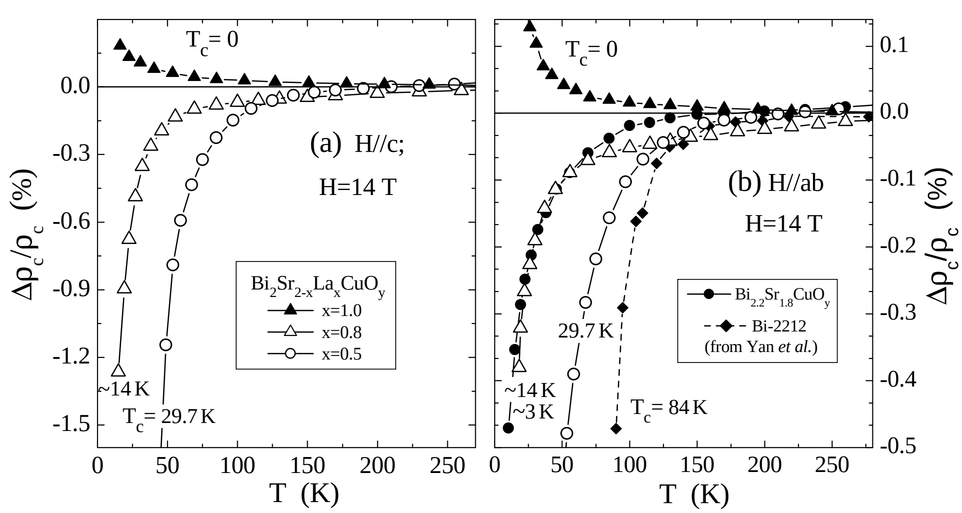

Fig. 4 – T dependences of the longitudinal (a) and transverse (b) c-axis MR at 14 T for BSLCO and BSCO crystals. The T_c values are indicated near each curve. The MR data for Bi-2212 from Yan *et al.* [14] are shown for comparison.

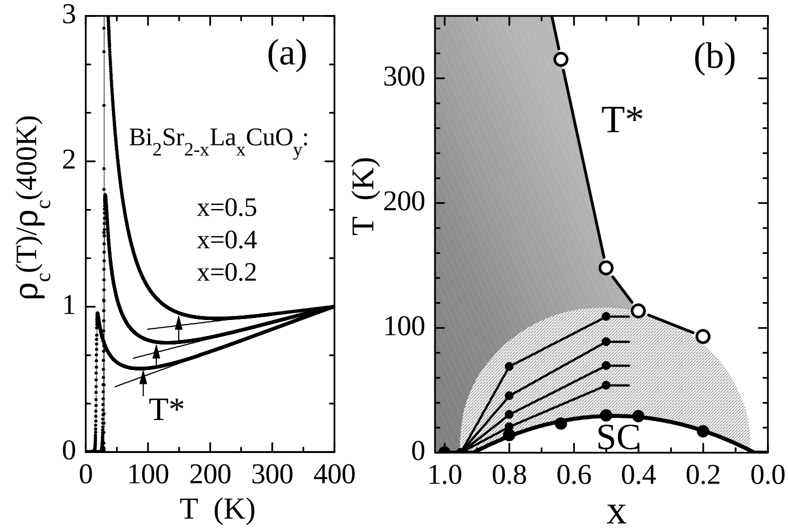

Fig. 5 – (a) c-axis resistivity of $Bi_2Sr_{2-x}La_xCuO_y$. T^* is defined as a temperature where $\rho_c(T)$ deviates from its linear extrapolation by 10%. (b) Schematic phase diagram of BSLCO, showing the PG region below T^* (shaded) and the region where the large negative MR is observed (hatched); a set of thin lines corresponds to the longitudinal MR levels of (from above) 0.1, 0.2, 0.4, and 0.8%.

sign of the MR appears to be determined by the low-temperature ground state. This indicates that the mechanism which causes the negative MR sets in already at such high temperatures as $\sim 150\,\mathrm{K}$ and that this mechanism only exists when the sample becomes superconducting at low temperatures.

Based on the behaviors of $\rho_c(T)$ and $\Delta\rho_c/\rho_c$, a schematic phase diagram of BSLCO is sketched in fig. 5, where T^* is defined as a temperature where $\rho_c(T)$ deviates from its linear high-temperature behavior by 10% [fig. 5(a)]. The evolution of T^* with increasing hole density turns out to be notably non-monotonic: a rapid change of T^* in the underdoped region is separated by a kink from a much slower decrease in the overdoped region. On the other hand, the hatched area in fig. 5(b), which marks the region where considerable negative MR is observed, follows T_c throughout the phase diagram, and apparently ignores the increase of T^* at low hole densities. For example, $|\Delta\rho_c/\rho_c|$ of the $x = 0.8$ sample (for which T^* is above $400\,\mathrm{K}$) stays less than 0.1% at $14\,\mathrm{T}$ in a wide temperature range below T^* and starts to grow rapidly only below $\sim 60\,\mathrm{K}$, while in the $x = 0.5$ sample (for which T^* is about $150\,\mathrm{K}$) a rapid growth of the negative MR is observed below $\sim 100\,\mathrm{K}$; this trend is irrespective of the field direction (fig. 4). A simple estimate shows that away from T_c the magnetic-field scale for suppression of the upturn in $\rho_c(T)$ (and thus of the normal-state PG) is extremely high: In the $x = 0.8$ sample, for instance, ρ_c increases by $\sim$3 times upon cooling from T^* to $150\,\mathrm{K}$, indicating that by $150\,\mathrm{K}$, $\sim$2/3 of the c-axis conductivity is wiped out by the PG. At the same time, an application of $H = 14\,\mathrm{T}$ at $150\,\mathrm{K}$ reduces ρ_c by only $\sim$0.04%; therefore, even if the MR kept the steep $\Delta\rho_c/\rho_c \propto H^2$ behavior up to the highest field, it would require $\sim 600\,\mathrm{T}$ to completely suppress the PG. It is worth noting that in $YBa_2Cu_3O_y$ the negative c-axis MR has also been found to disappear when SC is killed by underdoping [23].

The above result therefore distinguishes two essentially different pseudogapped regions on the underdoped side of the phase diagram: one is marked by the semiconducting $\rho_c(T)$ behavior and negligibly weak magnetic-field dependence; the other is marked by the SC-

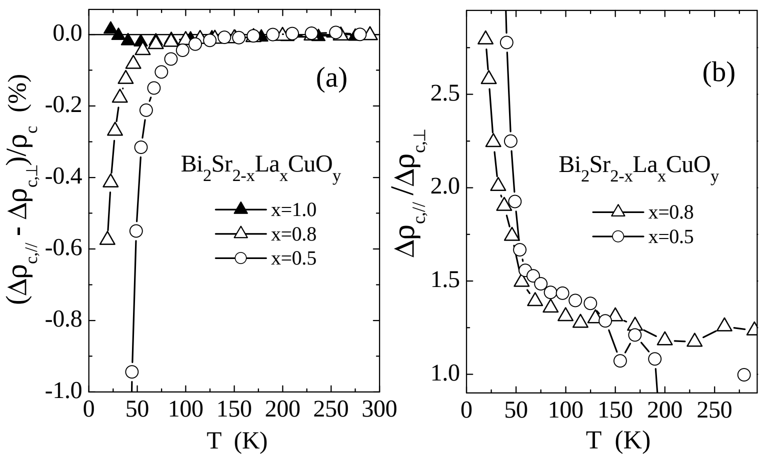

Fig. 6 – (a) Difference between the longitudinal and transverse MR obtained from the data in fig. 4. (b) The MR anisotropy $\Delta\rho_{c,\|}/\Delta\rho_{c,\perp}$.

related large negative MR. This distinction can be understood if one assumes that there are two distinct mechanisms that separately contribute to the PG behavior. The predominant one that sets in around T^* in underdoped samples is almost insensitive to the magnetic field, gains strength with decreasing hole concentration, and smoothly extends into the non-SC region. The second one exists only in samples that show superconductivity, causes further pseudogapping, and can be suppressed by the magnetic field. On the overdoped side of the phase diagram, the distinction between these two is not clear any more.

The first mechanism of the PG characterized by T^* can probably be associated with antiferromagnetic (AF) [24] or stripe [25] correlations that develop in the CuO_2 planes; these correlations are fundamentally governed by the AF interactions and gain strength as the hole density is reduced towards the AF region. Note that the large exchange coupling J ($\approx 0.12\,\mathrm{eV}$) makes AF interactions (and the PG caused thereby) robust against the magnetic field. On the other hand, the second PG mechanism can most easily be understood by attributing it to the SC fluctuations, which are magnetic-field sensitive. One may naturally expect that the PG features associated with SC fluctuations [19] would occur in the vicinity of the SC region, exhibit a singularity at T_c, and disappear in non-SC compositions – exactly the behavior that has been observed for the secondary PG.

In fact, useful information on the PG mechanisms can be obtained from the MR anisotropy shown in fig. 6. At $T > 140\,\mathrm{K}$, the MR is almost isotropic: difference between the longitudinal MR ($\Delta\rho_{c,\|}$ for $H \parallel c$) and transverse MR ($\Delta\rho_{c,\perp}$ for $H \perp c$) is virtually zero [fig. 6(a)] and thus the ratio $\Delta\rho_{c,\|}/\Delta\rho_{c,\perp}$ is nearly one [fig. 6(b)], which suggests that the spin terms dominate at high temperatures. At lower temperatures, however, the longitudinal MR significantly exceeds the transverse MR in the SC samples (but not in the non-SC one); this means that the magnetic field $H \parallel c$, which affects the *in-plane* motion of carriers, becomes more effective in suppressing the PG than $H \parallel ab$. This anisotropy may be readily understood if the MR originates from the suppression of two-dimensional SC fluctuations in CuO_2 planes [19], where the "orbital" effects of the magnetic field are relevant only for $H \parallel c$. It is intriguing that the temperature range where the large negative MR is observed appears to be notably broader than is usually expected for SC fluctuations: in $Bi_{2.2}Sr_{1.8}CuO_y$, it extends up to $\sim 100\,\mathrm{K}$, though T_c is only $\sim 3\,\mathrm{K}$. This corresponds nicely to the recent scanning tunneling spectroscopy

of the PG in overdoped BSCO [26], which also finds that the PG temperature is extremely high compared to T_c.

The existence of two distinct mechanisms, which both cause the depression of the DOS, might be the answer to the long-standing debates on whether the PG has a SC [2,3,19] or non-SC [8,9,12] origin: there appears to be both a non-SC pseudogap (that gains strength at low doping and tends to vanish in the overdoped region [9]) and a SC-induced suppression in the DOS (that persists in a certain range above T_c at all dopings [2,26]). The two mechanisms with contrasting doping and magnetic-field dependences seem to be the main source of confusion in studies of the PG in cuprates; measurements that probe the PG at high temperatures and low doping apparently see the magnetic-field insensitive part of the PG [11,12], while those performed at low temperatures and higher doping see the SC-related part of the PG that is sensitive to the magnetic field [10,13].

In summary, we present c-axis magnetotransport data of Bi-2201 that strongly suggest existence of two distinct mechanisms for the pseudogap in cuprates. The first one is almost insensitive to the magnetic field and causes the primary pseudogap opening at T^* in the underdoped region; we discuss this may be related to the development of the magnetic correlations or stripes. The second one is very magnetic-field sensitive, which is the source of the large negative MR, and causes a further pseudogapping as temperature is decreased towards T_c. This second mechanism is found to be present only in superconducting samples and is most strongly suppressed when the magnetic field is applied along the c-axis, which points to its relation to the superconductivity.

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